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Predicting the Visibility of Chimney Plumes
An Intercomparison of Four Models with
Observations at a Well-Controlled
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PREDICTING THE VISIBILITY OF CHIMNEY PLUMES: AN INTERCOMPARISON OF FOUR MODELS WITH OBSERVATIONS AT A WELL-CONTROLLED POWER PLANT

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Abstract—Four models for the operational prediction of plume visibility have been run by their developers on data from the VISTTA field study at the Navajo Generating Station. The different models give substantially different predictions of plume dispersion, cross-plume concentration integrals, and plume-sky contrast. Two of the models predict plume-sky contrasts which differ qualitatively from those observed. Overall model performance is limited chiefly by dispersion module performance; given observed concentration integrals as inputs, three of the models correctly predict the qualitative features of observed contrast. Much of the observed variation in plume dispersion and concentration integrals appears to have been due to sub-scale fluctuations in atmospheric transport and source emissions. The effects of particles on observed plume contrast were less than the experimental uncertainty.

Key word index: Visibility, plumes, dispersion, validation, nitrogen dioxide.

INTRODUCTION

In 1977, the U.S. Congress declared the conservation and restoration of visual air quality in certain scenic areas to be a national goal. The Environmental Protection Agency (EPA) responded (Costle, 1980) with a strategy which begins with the control of visible plumes in the vicinity of point sources. As a result of these developments, the permitting process for a major new plant or operation must now include an evaluation of the visual impact of its plume. Against this background, several groups have developed computer models designed to predict the quantitative effects of specified emissions on the optical environment.

To provide a basis for the evaluation of plume visibility models, the U.S. EPA and the Salt River Project (SRP) in 1979 cosponsored the VISTTA (Visibility Impairment due to Sulfur Transport and Transformation in the Atmosphere) program of intensive field measurements at SRP's Navajo Generating Station in northern Arizona. The present report presents results from a cooperative model intercomparison study based on these measurements. The intercomparison was sponsored by the U.S. EPA and the American Petroleum Institute (API), and included the four leading models for operational prediction of plume visibility. The results presented here address the accuracy and internal consistency of

the VISTTA data, the performance of individual models and submodules, and the considerations involved in comparing predictions with observations.

THE MODELS

The following models were evaluated in the Intercomparison Study.

ERT Visibility Model, Version 3. This model is offered by Environmental Research & Technology, Inc., and is fully documented (Drivas *et al.*, 1980). It has been described in the open literature (Drivas *et al.*, 1981), analyzed for sensitivity to inputs (Machiraju *et al.*, 1982), and compared with field data (Seigneur *et al.*, 1982).

Los Alamos Visibility Model. This model, developed with partial funding from the U.S. EPA, is offered by Los Alamos National Laboratory and is fully documented (Nochumson *et al.*, 1982). It has been described in the open literature (Williams *et al.*, 1980, 1981) and compared with two independent sets of field data (Williams *et al.*, 1981; Williams and Nochumson, 1982).

PHOENIX. This model is offered by Radian Corporation, with no documentation yet available. The dispersion, chemistry, and aerosol modules have been described in the open literature (Eltgroth and Hobbs, 1979), and the optics module in reports (Eltgroth, 1982; Hobbs *et al.*, 1982). The full model has been compared with field data (Eltgroth, 1982).

PLUVUE. This model, developed under contract to the U.S. EPA, is offered by Systems Applications, Inc., and is fully documented (Johnson *et al.*, 1980). It has been described in the open literature (Latimer and Samuelsen, 1978; Latimer, 1980), analyzed for sensitivity to inputs (Seigneur *et al.*, 1982; Machiraju *et al.*, 1982), and compared with two independent sets of field data (Bergstrom *et al.*, 1981; Seigneur *et al.*, 1982; Eltgroth, 1982).

The four models are similar in concept, all of them having the essentially linear structure indicated in Fig. 1. They differ significantly, however, in their emphasis and level of detail. The Los Alamos model incorporates the most explicit simulation of radiative transfer, and can give the most comprehensive description of plume appearance. The model PHOENIX includes the most explicit treatment of chemical reaction and

aerosol growth, and is the only one to base its dispersion calculation on eddy diffusivities rather than standard Gaussian parameters. The ERT model and PLUVUE are both substantially more parameterized, and correspondingly easier to run. Of the two, the ERT model is less demanding of computer resources and PLUVUE is more widely used and tested.

THE DATA BASE

The intercomparisons presented here are based on a cooperative field study, carried out as part of Project VISTTA, of the plume from the Navajo Generating Station in northern Arizona (Blumenthal *et al.*, 1981). Navajo is a modern plant, with three 750 mW units firing low-sulfur coal and equipped with efficient precipitators. It was chosen for study as representative of plants likely to be built in the future; it is less representative of plants already in existence (White and Patterson, 1981). The measurement program was specifically designed to provide data for the validation of plume visibility models. The core of the design was the use of instrumented aircraft to map the distribution of particles and trace gases along telephotometer sight paths (Richards *et al.*, 1981a; Bergstrom *et al.*, 1981). Supporting operations characterized plant emissions (Candelaria and Palomino, 1982) and the background air (Macias *et al.*, 1981; Hering *et al.*, 1981; Wilson and McMurry, 1981). Measurements were carried out during June, July and December of 1979, under conditions favorable to the formation of a visible plume.

The data for 27 sets of experimental observations, each comprising a telephotometer reading of plume/background contrast, an approximately coincident plume traverse, and supporting meteorological plant, and background air measurements, are summarized in the VISTTA Interim Report (Blumenthal *et al.*, 1982). Twenty two of these sets, taken on 28 June, 13 July, 7 December and 15 December were used in the Intercomparison Study for runs of the full models. In all but two of these 22 sets the plume was sighted against an unclouded sky. The two 13 July sets include aircraft data from a common traverse, and one of the telephotometer sightings on 28 June was against poorly-characterized terrain. The maximum number of distinct, valid data available for an individual

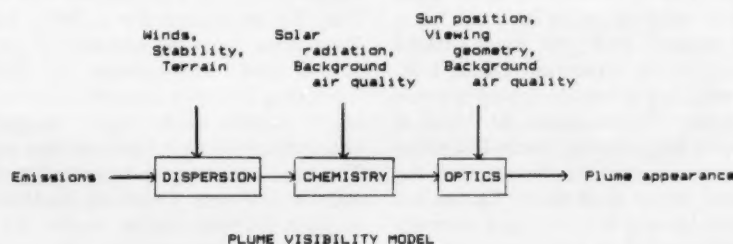


Fig. 1. The organization of modules in a plume visibility model.

parameter is thus 21, and the number of nitrogen dioxide data is 20 due to instrument overranging on one 15 December traverse. A subcollection of ten sets, all taken on 7 December against unclouded sky, was used for runs of the optics modules.

Selected characteristics of the 22-set data base are summarized in Table 1. It can be seen that all observations were made at moderate distances in a very clean environment. The plume was relatively poor in light-scattering particles, and was usually viewed away from the sun.

The performance of a model cannot reasonably be evaluated without some understanding of the uncertainties in the observations to which it is compared. These uncertainties arise both from random measurement error and, more fundamentally, from unpredictable short-term fluctuations in the actual plume. An idea of their combined magnitude can be obtained from comparisons of observations made under similar circumstances. Table 2 shows absolute differences recorded in the 10 sets of observations most closely paired by time and viewing geometry. The sample is small and the scatter large, so that summary statistics must be treated with caution. The observed differences are instructive, nevertheless, as direct indications of the baseline scatter which cannot be avoided in comparisons of models with observations. The median of the observed absolute differences will be taken in what follows as a conservative estimate for the aggregate nonsystematic uncertainty inherent in a single measurement; this includes the effects of real fluctuations in the atmosphere, not just measurement error. The values thus obtained for the uncertainty in

Table 1. Selected characteristics of the data base

Parameter	Minimum	Median	Maximum
Downwind distance of observation (km)	4	18	31
Range of observer from plume (km)	12	18	33
Visual range outside plume (km)	177	274	292
Ozone concentration outside plume (ppb)	31	35	44
Sun-sightpath angle (degrees)	26	140	159
NO _x /B _{scat} (550 nm), ratio of cross-plume integrals (ppm-km)	4	9	12

observed contrast are compatible with those derived by Blumenthal *et al.* (1982) from a closer examination of the raw data. The relative uncertainties in the aircraft measurements are larger than those in the telephotometer measurements, a possible consequence of operational difficulties in flying through the exact center of a strongly stratified plume.

STUDY PROTOCOL

Participating modelers were furnished with two levels of input data, one sufficient to run the full model and one sufficient to run the optics module by itself. For reasons of economy, the optics modules were run only on the subcollection of 10 observation sets earlier selected by the developers of PLUVUE (Bergstrom *et al.*, 1981). The data supplied are summarized in

Table 2. Comparisons of paired observations

Date		Paired observations						Medians	
		12/7	12/7	12/7	12/7	12/15	12/15		
Time of first observation (MST)		0908	1030	1427	1505	1125	1209		
Time of second observation (MST)		0929	1044	1438	1540	1133	1236		
Absolute differences within pairs (Δ value)								Value	Δ Value
Time and viewing geometry									
time of day	(min)	21	14	11	35	8	27	—	17
azimuth of sighting	(deg)	5.9	0.0	1.3	1.6	*	0.8	—	1.3
distance of observation from stack	(km)	0.0	0.7	0.5	0.5	2.2	0.3	21.3	0.5
distance of observer from plume	(km)	2.9	1.3	4.7	1.5	*	0.6	19.0	1.5
Aircraft measurements									
cross-plume NO ₂	(ppm-km)	0.107	0.019	0.022	0.339	>0.107	†	0.196	0.107
cross-plume B _{scat} at 550 nm	(—)	0.070	0.020	0.014	0.110	0.212	†	0.101	0.070
plume width at 20 ppb SO ₂	(km)	4.44	0.15	0.72	11.81	0.18	†	7.4	0.72
Telephotometer measurements									
contrast at 450 nm	(—)	0.021	0.023	0.008	0.163	*	0.009	0.143	0.021
contrast at 630 nm	(—)	0.004	0.015	0.006	0.024	*	0.007	0.024	0.007

The body of the table gives absolute differences within pairs. Right-hand columns give medians of the 10 or 12 individual values (V) and 5 or 6 pairwise differences (Δ V). *Telephotometer sight paths were not aligned.

† Aircraft traverses were not aligned.

Table 3, and were taken to be as given by the VISTTA Interim Report (Blumenthal *et al.*, 1982). The VISTTA Report was coauthored by the developers of PLUVUE (who participated in the design and execution of the field study), and contains additional data either synthesized by PLUVUE or assumed as input in its runs. Secondary data of this sort were available to the other modelers, but were not formally specified. The VISTTA Report also contains the observed values of the quantities to be predicted, but these were not used to tune the models.

The four models evaluated differ in their requirements and options for user-supplied data, and each requires some inputs which were not directly measured in the field. The judgement of its operator is thus an integral component of each model's performance in the Intercomparison Study, just as it would be in operational use. This is illustrated in Fig. 2, which compares predictions obtained by two different operators from the same model.

The PLUVUE runs used in this intercomparison were carried out as part of Project VISTTA (Bergstrom *et al.*, 1981; Seigneur *et al.*, 1982). The PLUVUE code has since been modified; its internal calculation for the solar zenith and scattering angles has been corrected, light absorption by carbonaceous

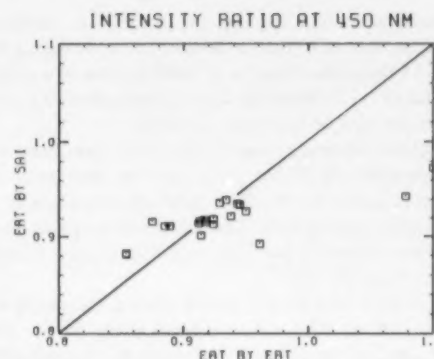


Fig. 2. Effects of operator discretion on model output. The ERT Visibility Model was run on the same VISTTA database by Environmental Research and Technology (this study) and Systems Applications, Inc. (Seigneur *et al.*, 1982). The resulting predictions of plume/background intensity ratios are not identical, as shown here by scatter about the diagonal of agreement.

aerosols has been incorporated, and its treatment of the diffuse radiation field has been refined. The ERT Visibility Model and PHOENIX were both run specifically for the present intercomparison study, the ERT

Table 3. Data supplied to model operators

Parameter	Basic data set for full models	Nominally independent measurements
Emissions		
SO ₂		3
NO _x		3
Particulate matter		3
Bimodal particle size distribution		1
O ₂ concentration		3
Exhaust characteristics		3
Geometry		
Coordinates (3D) of observer		21
Coordinates (3D) of plant		1
Stack height		1
Time of day		22
Direction of plume		22
Distance of plume observation from stack		22
Distance of observer from plume		21
Coordinates (2D) of sight path		21
Angle to plume centerline		21
Visual background		21
Environment		
Conditions at plume level		
Wind speed		6
Temperature		21
Relative humidity		21
Scattering coefficient at 550 nm		5
Unimodal particle size distribution		1
O ₂ concentration		21
Temperature profile		5
Opaque sky cover		5
Supplementary data set for optics modules		
Cross-plume integrals		
NO ₂ concentration		20
Particle scattering at 550 nm		21

model on all 22 observation sets and PHOENIX on the 18 from 7 December and 15 December. The ERT model was run with a user-supplied scattering phase function, generated outside the model from the VISTTA size distribution parameters. The Los Alamos model runs were carried out as part of an independent study (Williams and Nochumson, 1982), and were confined to the 10 observation sets selected for inter-comparison of optics modules.

INTERCOMPARISON OF THE OPTICS MODULES

Figures 3 and 4 display observed and predicted plume/sky intensity ratios for sight paths through, but not generally perpendicular to, the plume axis. The observations represent ten sightings on 7 December 1979, all against unclouded sky and away from the sun. The predictions are based on observed, rather than predicted, cross-plume integrals of B_{scat} and NO_2 . Figures 3 and 4 thus isolate the performance of the models' optics modules on a selected subset of the VISTTA data.

Figure 3 shows the performance of the optics modules at the blue end of the spectrum, where

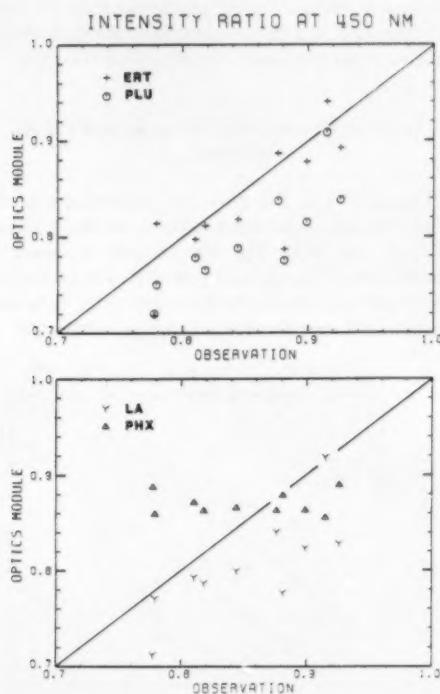


Fig. 3. Predicted and observed plume/background intensity ratios at 450 nm. Predictions are those of the optics submodules, based on observed cross-plume integrals. Models are ERT, ERT Visibility Model; PLU, PLUVUE; LA, Los Alamos Visibility Model; PHX, PHOENIX. Sloping line is the locus of model-observation agreement.

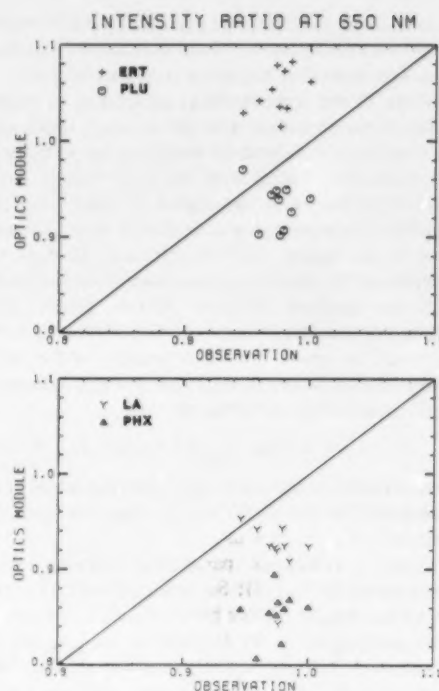


Fig. 4. Predicted and observed plume/background intensity ratios at 630 nm or 650 nm. Predictions are those of the optics submodules, based on cross-plume integrals.

observed plume/sky contrasts were greatest. Nitrogen dioxide absorbs strongly at 450 nm, and Richards *et al.* (1981a) concluded from their aircraft measurements that it was the dominant cause of extinction of blue light. The optics modules of ERT, PLUVUE and Los Alamos yield predictions which agree reasonably well with observation. This correlation of prediction with observation supports both the adequacy of these models' radiative transfer treatments and the cross-consistency of the aircraft and telephotometer data. The optics module of PHOENIX, in contrast, yields predictions which are unresponsive to variations in the B_{scat} and NO_2 inputs, and do not correlate with observation. The reason for PHOENIX's behaviour is not known.

Figure 4 compares predictions of the optics modules at 650 nm or 630 nm with telephotometer observations at 630 nm. Observed plume/sky contrasts were small at the red end of the spectrum, where absorption by nitrogen dioxide is minor and extinction is controlled by particle scattering (Richards *et al.*, 1981a). The predictions of the several models diverge significantly, presumably due to differing estimates of particle albedo and phase function. Errors in the treatment of the plume's inherent optical characteristics distort predicted contrasts most strongly in the red, because the near-Rayleigh atmosphere between plume and observer is most transparent here. Such errors may

have arisen in part from the particle size distributions specified as model inputs, which were based on incomplete and somewhat uncertain experimental data.

Much of the computational complexity of plume visibility models is associated with the need to account for directionally dependent scattering by particles in the plume and background air. It is thus of some interest to determine the degree to which particles contribute to observed and predicted plume appearance in the present data set. This can be done by comparing the observed and simulated Navajo plume with an idealized nitrogen dioxide plume. The plume/sky contrast of this particle-free plume can be expressed in closed form as a function of the NO_2 integral along the line of sight and the optical distance of the plume from the observer:

$$(I_s - I_p)/I_s = \exp(-\tau_{\text{air}})[1 - \exp(-\tau_{\text{NO}_2})]. \quad (1)$$

The derivation of this relationship, and the details of its evaluation for the VISTTA data base, are given in Appendix A.

Figure 5 compares the 450 nm intensity ratios predicted by the NO_2 reference calculation (1) and the optics modules of ERT, PLUVUE, and Los Alamos. (The predictions of PHOENIX, which are uncorrelated with nitrogen dioxide, are not shown.) The predictions of all three optics modules are consistently close to those of the NO_2 reference, the predictions of PLUVUE and Los Alamos exhibiting a nearly constant offset. (The magnitude of the offset for PLUVUE is consistent with the results of an earlier sensitivity analysis (Seigneur *et al.*, 1982), which found that a 20% decrease in particle emissions produced a 0.6% increase in the predicted intensity ratio at 450 nm.) It is apparent that the 450 nm intensity ratio predicted by the models is controlled by NO_2 absorption. Moreover, a comparison with observation shows that the inclusion of particle optics in the modules does not improve their performance over that of the simple NO_2 reference calculation.

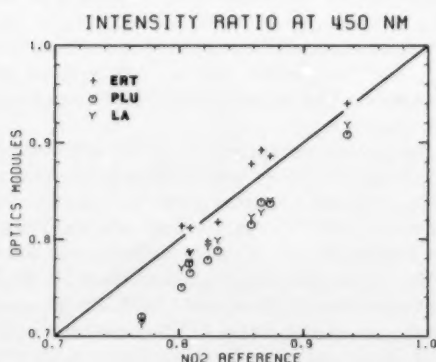


Fig. 5. Predicted effects of particle scattering and absorption at 450 nm. Intensity ratios predicted by optics submodules are compared with those expected of a particle-free NO_2 plume.

Figure 6 compares observed 450 nm intensity ratios with those predicted by the NO_2 reference calculation (1). The observed and predicted values correlate moderately well, exhibiting better agreement for the (circled) sightings selected for optics module runs than for the remainder of the data set. Since all but 2 of the 20 sightings were taken with the sun at the observer's back, the effect of plume particles should generally have been to lower the observed intensity ratio (White and Patterson, 1981). The absence of a systematic difference between observation and NO_2 reference calculation is thus consistent with a conclusion that the observed 450 nm intensity ratio is accounted for by NO_2 absorption. (The observed 630 nm intensity ratios of almost 1.0 in Fig. 4 can also be accounted for by NO_2 absorption, which is insignificant at this wavelength.)

The source of the scatter in Fig. 6 has not been identified. If particle extinction and scattering are truly negligible, then the discrepancies between Equation (1) and observation must be due to errors in the telephotometer measurement, the aircraft measurement, or the alignment of the two. The individual telephotometer and aircraft measurements of the plume are relatively accurate. The alignment of sighting and traverse, on the other hand, is a potential source of significant error, as is the characterization of background optics by a single number distilled from all data taken outside the plume during several traverses.

INTERCOMPARISON OF THE REACTIVE PLUME SUBMODELS

Figures 7 and 8 display the performance of the plume models with respect to the crosswind integrals of B_{scat} and NO_2 . The observations represent 21 distinct aircraft traverses on four days. The predictions are based on available data for wind speed, lapse rate, background concentrations, and plant emissions.

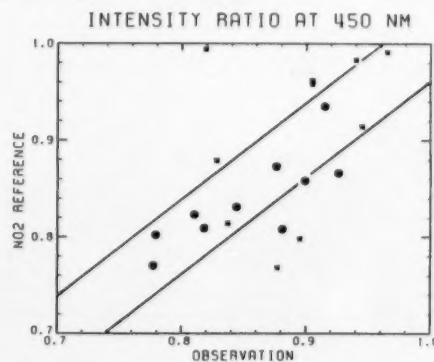


Fig. 6. Comparison of telephotometer and aircraft data. Intensity ratio measured by telephotometer is plotted against intensity ratio calculated from nitrogen dioxide measured by aircraft (Appendix A). Half of the points lie within the sloping lines. Sightings selected for optics module runs are circled.

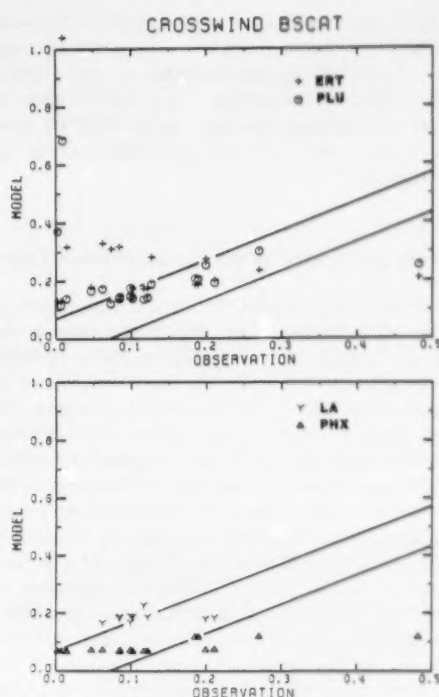


Fig. 7. Predicted and observed cross-plume integrals of particle scattering at 550 nm. Sloping lines indicate inherent uncertainty of model-observation comparison (cf. Table 2).

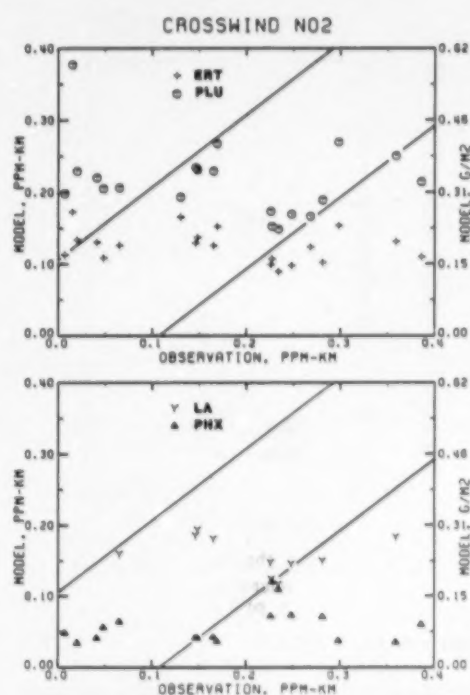


Fig. 8. Predicted and observed cross-plume integrals of nitrogen dioxide concentration. Predictions are in g m^{-2} for the ERT and Los Alamos models, ppm-km PLUVUE and PHOENIX.

Figure 7 shows observed and predicted values of the extinction at 550 nm due to scattering by plume particles. The quantity plotted is the (dimensionless) integral of the particle scattering coefficient (B_{scat}), along a line through a perpendicular to the plume axis. (Observed values have been adjusted for oblique flight paths.) Sloping lines indicate the region in which models and observations agree to within experimental uncertainty, estimated from the paired observations of Table 2. The model predictions generally exhibit less variability than the observations, except for a single traverse appearing in the upper left corner. In this one instance, the first of two 28 June traverses, the predictions of ERT and PLUVUE (the only models run) are very high and the observed value is low. The judgement of the field crew at the time was that the aircraft might have missed the plume center on this occasion.

Figure 8 compares observed and predicted values of the nitrogen dioxide concentration. The quantity plotted is the NO_2 analog of the B_{scat} integral plotted in Fig. 7. Observations, and the predictions of PLUVUE and PHOENIX, were reported in ppm-km, while the predictions of the ERT and Los Alamos models were reported in g m^{-2} . All four sets of predictions are plotted as received, in their original units, on separate scales which agree for conditions (at plume level) of

0.785 atm and 10°C . The scales agree to within 2% for most, and 8% for all, of the points plotted. The variability in the predictions of any one model is less than the variability in the observations. There is more variability among the predictions of different models, with those of PLUVUE and PHOENIX respectively the highest and lowest of the lot.

It is evident from Figs 7 and 8 that there is little correlation between observation and prediction for the B_{scat} and NO_2 integrals. This disappointing result does not imply that there is any fundamental error in the models. All that can be concluded from this absence of correlation is that the uncertainty in the model predictions is large relative to the range of predictions, as illustrated in Fig. 9. Figure 9 plots the NO_2 residual (observation minus prediction) as a function of prediction, for the two models which were run on all 20 data sets. (One outlier, corresponding to the 28 June traverse noted above, is not plotted.) There is no correlation between residual and prediction, which means that the performance of these models over the range of conditions tested can be described by an expression of the form

$$X_o = X_p + E,$$

where X_o and X_p are the observed and predicted values and E is a random error independent of the

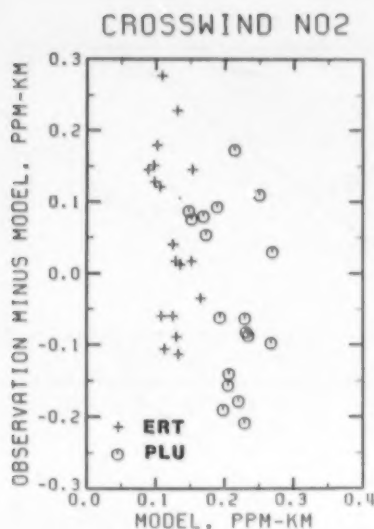


Fig. 9. Independence of NO₂ residual and prediction.

prediction. The median absolute scatter in the NO₂ error terms, defined as one-half the difference between their 75 and 25 percentile values, is 0.110 ppm-km for PLUVUE and 0.103 ppm-km for the ERT model. This is consistent with the earlier estimate of 0.107 ppm-km for the uncertainty inherent in the comparison of individual observations and predictions, based on the paired observations of Table 2.

The factors contributing to discrepancies between prediction and observation include errors in the theoretical formulation and computational implementation of the models, fluctuations in emissions and atmospheric transport below the spatial and temporal scales resolvable by the models, errors in the inputs specified to the models, and errors in the experimental observations. Only the first three of these are inherent in the modeling process, and it is thus of some interest to determine the importance of the remaining factor, experimental error. Figures 3 and 6 show that the NO₂ measurements are consistent with telephotometer measurements made independently. More precisely, the median absolute difference between the 450 nm intensity ratios observed and predicted from observed NO₂ integrals is 0.04, as indicated by sloping lines in Fig. 6. A standard analysis of error propagation yields from this value an upper bound of 0.04 ppm-km for the median absolute error in the measurement of the NO₂ integral (Appendix B). The majority of the 0.10–0.11 ppm-km apparent error in the NO₂ predictions of the reactive plume models is thus real, and must be attributed to limitations in model formulation, resolution, and input.

The bulk of the error in NO₂ predictions appears to be random rather than systematic, indicating that it is attributable to sub-scale fluctuations in the actual plume rather than to errors in model formulation or

inputs. It would thus be of some interest to compare predictions with the averages of observations made under conditions indistinguishable to the models. Unfortunately, it is clear from Fig. 9 that the range of observing conditions available in the VISTTA data base is too small for such a comparison to be meaningful.

INTERCOMPARISON OF THE DISPERSION MODULES

The treatment of dispersion is a critical aspect of plume visibility modeling. The time-averaging involved in human vision is negligible, and a visibility model must therefore attempt to predict instantaneous concentration fields. This is a relatively new goal, one which did not figure in the evolution of the familiar parametrizations routinely used for plume dispersion.

Of the two crosswind components of dispersion, the vertical is the more important in determining plume appearance, since it controls the quantity of material along the near-horizontal line of sight of a distant observer. Unfortunately, VISTTA's experimental design did not yield direct observations of the plume's vertical dimension. An indication of model performance on this parameter is, however, available in the results for cross-plume B_{scat} integrals (Fig. 7). Light-scattering aerosol was a conserved species at the downwind distances under consideration (Richards *et al.*, 1981a), so the cross-plume B_{scat} integral should vary inversely with vertical dispersion. The failure of the models to account for observed variations in this integral is thus suggestive of an inability to predict variations in vertical dispersion. (It is not conclusive evidence, because part of the variability in the B_{scat} integral is due to significant fluctuations in flyash emission rates (Richards *et al.*, 1981a).)

Horizontal dispersion is important chiefly for its contribution to the entrainment of background ozone, the limiting reactant in the near-field conversion of NO to NO₂. At the downwind distances under consideration, partitioning of the nitrogen oxides is controlled by the fast photolytic cycle:



All three of these reactions are sufficiently fast that the system should be near photostationary state throughout most of the plume, with reactant concentrations determined by local dilution. Figure 10 shows cross-plume NO₂ integrals calculated on this basis for a Gaussian plume having both maximum and integrated NO_x concentrations equal to those observed (Appendix C). It is evident from the correlation between calculated and observed values that we can predict the chemical conversion of NO to NO₂ if we can predict the physical dispersion of NO_x.

Although VISTTA characterized horizontal plume

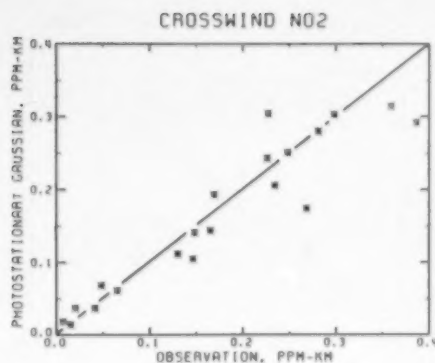


Fig. 10. Simplicity of observed NO_2 chemistry. Observed NO_2 integral is plotted against NO_2 integral expected from reactions R1-R3 on the basis of observed NO_2 integral, NO_2 maximum, and O_3 background (Appendix C).

dispersion rather well, direct comparison of observation and prediction is attended by some ambiguity. All four dispersion modules predict Gaussian distributions of effluent in the horizontal, so that lateral dispersion is completely described by the single parameter σ_y . The concentration profiles recorded in aircraft traverses were, on the other hand, often strongly non-Gaussian, indicating interleaved regions of high and low effluent concentrations (Richards *et al.*, 1980, 1981b). The Gaussian dispersion parameters corresponding to the observed profiles can be defined in different ways; to the degree that the profiles are non-Gaussian, these definitions can yield different values. Such disagreement is illustrated in Fig. 11(a), which shows as a function of downwind distance two different definitions of the observed dispersion parameter. The parameter SY (50% max), a measure of the geometric spread of the plume, is derived from the width $2y_{0.5}$ of the plume at a concentration level one-half of maximum; it is defined as the value of σ_y determined by the relationship $\exp(-y_{0.5}^2/2\sigma_y^2) = 0.5$. The parameter SY (max, int), a measure of plume dilution, is derived from the cross-plume integral and maximum value for the traverse; it is defined as the value of σ_y determined by the relationship $\int [\text{NO}_x] dy = \sqrt{2\pi} \sigma_y [\text{NO}_x]_{\text{max}}$.

Figure 11(b) shows the Gaussian parameters σ_y predicted by the four dispersion modules. The operators of the ERT model and PLUVUE both opted to use the standard Pasquill-Gifford curves (Turner, 1970) for stability classes D (neutral) and E (stable). The Los Alamos model was run with the curve developed by TVA (Montgomery *et al.*, 1973) for very stable conditions. The vertical profiles of eddy diffusivities required by PHOENIX were taken from a plot for stable conditions reproduced by Pasquill (1974). The various predictive schemes differ substantially, and these differences propagate through the chemistry and optics modules.

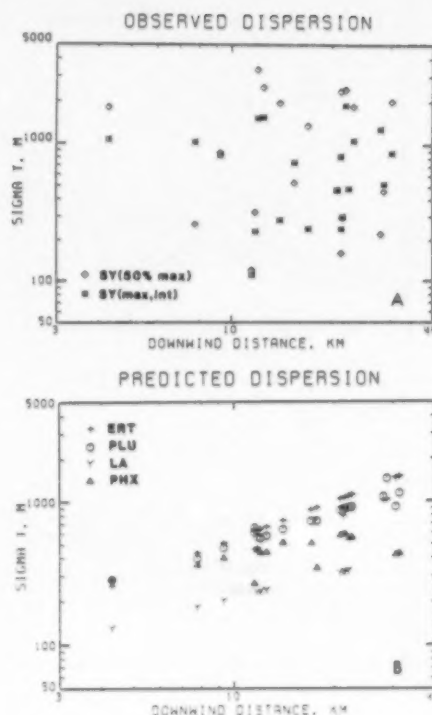


Fig. 11. Two measures of observed horizontal dispersion, and Gaussian parameters predicted by the models.

Figure 11(b) highlights an important consideration in the evaluation of model performance. The dispersion modules of ERT, Los Alamos and PLUVUE are all based on a common collection of standard parametrizations. Differences in their numerical implementation of these parametrizations are minor, so that the three dispersion modules can be considered as roughly equivalent pieces of computer code. The predictions of the three modules differed because the operator of the Los Alamos model differed from the others in his selection of parametrization, and because the operators of the ERT model and PLUVUE differed with each other in their typing of stability. Figure 11(b), like Fig. 2, shows that it is misleading to view a model as an inert piece of computer code; in operational use, a model is an amalgam of deterministic algorithm and flexible operator judgement.

INTERCOMPARISON OF THE INTEGRATED MODELS

Figure 12 shows the 450 nm intensity ratios observed and predicted by the plume visibility models under conditions simulating operational use. The observations represent 21 distinct telephotometer scans on four days. The predictions are based on available data for winds, stability, background air quality, emissions, and viewing geometry.

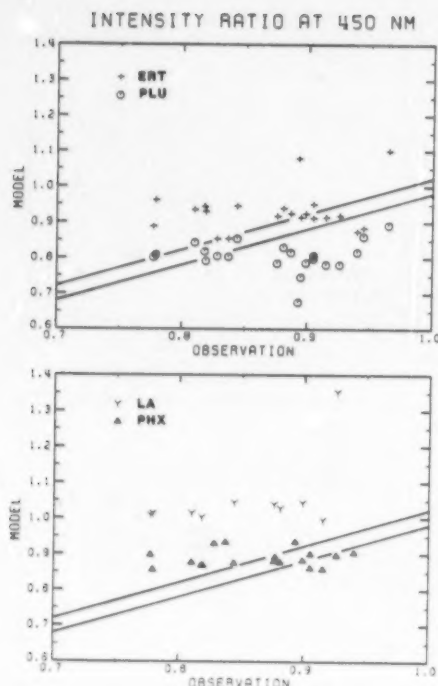


Fig. 12. Predicted and observed plume/background intensity ratios at 450 nm. Predictions are those of the full models, so that optics calculations are based on predicted cross-plume integrals shown in Figs 7 and 8.

The four models yield significantly differing predictions of plume appearance. At one end of the range is PLUVUE, which tends to predict the darkest plumes. This behaviour is qualitatively consistent with PLUVUE's tendency to predict the largest NO_2 integrals. At the other end of the range is the Los Alamos model, which tends to predict plumes brighter at all wavelengths than the background sky. The brightness of the Los Alamos plumes is surprising, and is not understood. The excess plumelight can only come from particles, but our examination of the Los Alamos optics module shows the apparent effect of particles to be instead a slight darkening of the predicted plume (Fig. 5). The latter effect is what would be expected (White and Patterson, 1981) for the range of scattering angles (120° – 160°) covered by the Los Alamos runs.

The analyses in the preceding sections suggest that the performance of the integrated models is limited ultimately by the performance of their dispersion modules. Indeed, even a highly idealized model can reproduce the gross features of observed plume/sky contrast if it is supplied with the correct physical dispersion of NO_x as an input. This is illustrated in Fig. 13, which shows intensity ratios calculated for a particle-free Gaussian plume, in photostationary state, having maximum and integrated NO_x concentrations equal to those observed. (Figure 13 combines the

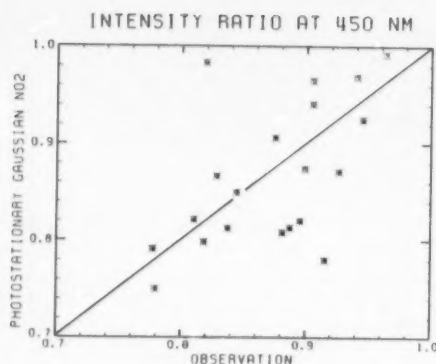


Fig. 13. Intensity ratios calculated for a Gaussian NO_x plume with observed physical dispersion. Simple calculations are those of Figs 6 and 10, based on the observed NO_x integral, NO_x maximum, and O_3 background.

simple calculations underlying Figs 6 and 10.) The correlation between observed and calculated values is 0.71 if a single outlier is deleted.

DISCUSSION

Given the regulatory importance attached to simulation modeling, the theoretical basis for the validation of models has so far received relatively little attention. Work to date (e.g. Fox, 1981; Willmott, 1982) has tended to focus on the statistical considerations involved in characterizing model performance on a given data base. A model's performance on one data set is an uncertain guide to its performance on another data set, however, and it is in such extrapolation to new conditions that models are most frequently applied. A full description of model validity must, therefore, include a complete characterization of the data set on which statistical measures of model performance are based.

The effects of data selection on model performance can be illustrated without even leaving the setting of the VISTTA measurements. An important characteristic of the data set employed here is its bias toward clearly visible plumes. This bias limits the range of both model inputs, such as stability class, and outputs, such as intensity ratio. Although the experimental and regulatory rationales for such a bias are obvious, it is instructive to imagine the results of a differently designed measurement program. Our imagined measurements would be similar to those of Project VISTTA, except that half of them would be vacuous—made under strongly unstable conditions, say, or with the observer 70 km from the plume on a hazy day. It seems reasonable to expect that models and observation would each yield intensity ratios near unity for these empty 'sightings'. The inclusion of the new data would then increase the correlation coefficient between model and observation, since it would increase the

variance of the predictions while decreasing the mean error of the predictions. The correlation between predicted and observed 450 nm intensity ratios would, for example, go from $r = 0.05$ to $r = 0.83$ for PLUVUE and from $r = 0.24$ to $r = 0.64$ for the ERT model. The addition to the data set of trivial observations carrying no physical information can, thus, sharply increase one common measure of model performance. It is to avoid this sort of ambiguity that we have relied so heavily on scatter plots for the presentation of our results.

The comparatively casual design of the present intercomparison limits its utility as a basis for model validation. The analysis presented here has, nevertheless, illustrated and explored two techniques which should be useful in a formal validation study. The first of these is the use of idealized but physically sound models in the characterization of the data set. The scatter between analytical models and observation can be simply translated into bounds in experimental error, as in Fig. 6 and Appendix B. The agreement with observation of a model which neglects certain effects establishes limitations in the coverage of the data, such as the unimportance of particle extinction (Fig. 6) or radical chemistry (Fig. 10). The second technique worth emphasis is the disaggregation of a model into independently testable modules. If, for example, a complex model requires 12 independent parameters as input, the number of observations needed to test its qualitative response to each parameter is at least $2^{12} = 4096$. If, on the other hand, it is possible to decompose this model into three independent modules each requiring four inputs, the minimum number of observations needed drops to $3 \times 2^4 = 48$.

CONCLUSIONS

A number of results have been presented here. The major conclusions regarding the VISTTA data set for the Navajo plume can be summarized as follows.

(i) The aircraft and telephotometer data taken in Project VISTTA are internally consistent. These data are sufficiently accurate that differences between models and observation cannot be attributed to experimental error.

(ii) The plume contrasts observed at the Navajo Generating Station can be predicted with reasonable success from the observed physical dispersion of NO_x . The effects of particles on observed plume contrast are less than the experimental uncertainty.

The comparison of model predictions with the VISTTA observations served as a useful test of model performance under a range of conditions relevant to potential applications. Although the intercomparisons undertaken were too informal and limited in scope to serve as positive validation, they did suffice to identify some specific limitations and problems.

(iii) Much of the variation observed in the Navajo plume appears to have been due to fluctuations in

source emissions and atmospheric transport below the spatial and temporal scales resolvable by the models. These fluctuations must be taken into account in projections of visibility impact, which is a function of the instantaneous plume.

(iv) Two of the four models tested fail to reproduce some qualitative feature of observed plume contrasts. The intensity ratios predicted from observed NO_2 and B_{scat} integrals by PHOENIX do not correlate with observation at 450 nm and, unlike observation, do not depend strongly on wavelength. The intensity ratios predicted from observed integrals by the Los Alamos model are in reasonable agreement with observation, but the intensity ratios predicted directly from emissions indicate a plume which is brighter than the sky at all wavelengths, unlike the dark plumes actually observed.

The study also yielded conclusions of a more general nature, pertaining to the methodological questions which arise in evaluating any air quality model.

(v) A description of model validity requires both a statistical measure of model performance and a characterization of the data set on which this statistical measure is based. For example, control of particulate emissions at the Navajo plant is sufficiently complete that the effects of particles on plume appearance cannot be identified in the VISTTA data. Validation of a model against data for the Navajo plume would not, therefore, insure satisfactory performance in a situation with higher particle loadings.

(vi) Model performance depends on operator judgement. The VISTTA measurements were designed to provide a data base much more comprehensive than any available to predictive models in operational use. Even so, all four models tested require input data which were not directly measured and had, therefore, to be assigned at the discretion of the operator. Identifiable differences in operator judgement contributed to significant differences in model outputs.

(vii) The evaluation of complex models with limited observations can be improved through the techniques of idealization and disaggregation. Idealized physical models provide an internal test of data consistency and coverage, thereby sharpening the interpretation of model-observation comparisons. Such a model for the plume/sky contrast due to nitrogen dioxide alone was used in this study to establish both the validity of the field data and the relative unimportance of particle scattering in the Navajo plume. Disaggregation of a model into independently testable modules greatly reduces the number of independent observations required for the mapping of its behaviour. Isolation of the optics module in this study made it possible to characterize its performance in a manner unaffected by the observed or modeled dispersion of the plume.

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APPENDIX A: DERIVATION OF EQUATION (1)

The equation governing radiative transfer along the observer's line of sight is (Chandrasekhar, 1960)

$$dI/dr = B_{\text{ext}} I - A,$$

where r is distance from the observer, I is intensity in the direction toward the observer and A is airlight—light scattered toward the observer by the atmosphere. In clean air, the airlight term is an approximately constant multiple of the extinction coefficient:

$$A = JB_{\text{ext}}.$$

Consider a particle-free plume of NO_2 viewed against unclouded sky. Let I_s^0 and I_p^0 be the intensity of the sky as viewed from, respectively, the far and near sides of the plume. The unobstructed sky intensity is given by

$$I_s^0 = (1 - e^{-\tau_0}) J,$$

where

$$\tau_0 = \int_{r_0}^{\infty} B_{\text{air}} dr$$

is the optical depth of the atmosphere beyond the plume, given by the integral of background extinction along the sightpath. The NO_2 plume filters this skylight, yielding a diminished intensity

$$I_p^0 = e^{-\tau_{\text{NO}_2}} I_s^0,$$

where

$$\tau_{\text{NO}_2} = \int B_{\text{NO}_2} dr$$

is the optical depth of plume NO_2 , given by the integral of NO_2 absorption along the sightpath. For an observer some distance from the plume, the intensity of the sky is given by

$$I_s = e^{-\tau_{\text{air}}} I_s^0 + (1 - e^{-\tau_{\text{air}}}) J,$$

and the intensity of the plume by

$$I_p = e^{-\tau_{\text{air}}} I_p^0 + (1 - e^{-\tau_{\text{air}}}) J,$$

where

$$\tau_{\text{air}} = \int_0^{r_0} B_{\text{air}} dr$$

is the optical depth of the air between observer and plume.

The contrast between plume and sky can be written

$$\frac{I_p - I_p^0}{I_s} = \frac{e^{-\tau_{\text{air}}} I_p^0 (1 - e^{-\tau_{\text{NO}_2}})}{e^{-\tau_{\text{air}}} I_s^0 + (1 - e^{-\tau_{\text{air}}}) J}.$$

At the low elevation angles (≤ 2 degrees) of the sightpaths treated in this study, the slant optical depth of the Rayleigh atmosphere exceeds 4 at 450 nm, so that $e^{-\tau_0} \approx 0$ and $I_s^0 \approx J$. The above expression for contrast then simplifies to Equation (1)

$$\frac{I_p - I_p^0}{I_s} = e^{-\tau_{\text{air}}} (1 - e^{-\tau_{\text{NO}_2}}).$$

In the calculations for Figs 6 and 13, the optical depth of plume NO_2 was derived from the NO_2 integral

$$\tau_{\text{NO}_2} = 1.355 (\sin \theta)^{-1} \int [\text{NO}_2] dy,$$

where $1.355 \text{ ppm}^{-1} \text{ km}^{-1}$ is the specific absorption of NO_2 at 450 nm (Hodkinson, 1966), θ is the angle between sightpath and plume centerline and y is crosswind distance. The optical depth of the air between observer and plume was derived

from the background scattering coefficient:

$$\tau_{\text{air}} = r B_{\text{scat}},$$

where r is the distance between observer and plume. The blue scattering coefficient was derived from the reported green scattering coefficient,

$$B_{\text{scat}}(b) = (538/450)^4 B_{\text{scat}}(g),$$

assuming a Rayleigh dependence on wavelength.

APPENDIX B: ANALYSIS OF ERROR PROPAGATION IN EQUATION (1)

Let s_X denote the median absolute error in X resulting from measurement errors. Let R denote the intensity ratio calculated from Equation (1). Under certain standard assumptions (Bevington, 1969), the error in R resulting from errors in the measurement of τ_{air} and $\int [\text{NO}_2] dy$ can be expressed as follows:

$$s_R^2 = \left(\frac{\partial R}{\partial \tau_{\text{air}}} s_{\tau_{\text{air}}} \right)^2 + \left(\frac{\partial R}{\partial \int [\text{NO}_2] dy} s_{\int [\text{NO}_2] dy} \right)^2. \quad (\text{B1})$$

The major assumptions underlying this expression are that the errors in τ_{air} and $\int [\text{NO}_2] dy$ are independent, which is a reasonable expectation in the present context, and small, which is somewhat less tenable. Errors in the sightpath-centerline angle and the specific absorption of NO_2 are neglected.

From Appendix A, the calculated intensity ratio has the following dependence on τ_{air} and $\int [\text{NO}_2] dy$

$$R = 1 - e^{-\tau_{\text{air}}} (1 - e^{-1.355(\sin \theta)^{-1} \int [\text{NO}_2] dy}).$$

The partial derivatives appearing in (B1) can be evaluated accordingly

$$\partial R / \partial \tau_{\text{air}} = 1 - R = 0.14$$

and

$$\begin{aligned} \partial R / \partial \int [\text{NO}_2] dy &= -1.355 (\sin \theta)^{-1} e^{-\tau_{\text{air}}} - 1.355 (\sin \theta)^{-1} \int [\text{NO}_2] dy \\ &= -0.97 \text{ ppm}^{-1} \text{ km}^{-1}. \end{aligned}$$

the numerical values being the medians for the data set. Seigneur *et al.* (1982) estimate the uncertainty in the characterization of background scattering to be about 25%, and we shall extend this estimate to τ_{air}

$$s_{\tau_{\text{air}}} = 0.25 \tau_{\text{air}} = 0.064.$$

Finally, the experimental error s_R in the calculated intensity ratio R is bounded by the (median absolute) difference between R and the observed intensity ratio I_p/I_s , shown in Fig. 6 to be 0.04. (This difference incorporates, in addition to the experimental error s_R , the unrelated error s_{I_p/I_s} in the measurement of the intensity ratio, and the unrelated error s_{model} resulting from the oversimple formulation of the model. From the presumptive independence of the several errors, it follows that

$$(0.04)^2 = [(I_p/I_s) - R]^2 = s_R^2 + s_{I_p/I_s}^2 + s_{\text{model}}^2 \geq s_R^2.$$

Substituting the values of the preceding paragraph into (B1) yields the following inequality

$$\begin{aligned} (0.04)^2 &\geq (0.14 \times 0.064)^2 \\ &\quad + (-0.97 \text{ ppm}^{-1} \text{ km}^{-1} \times s_{\int [\text{NO}_2] dy})^2. \end{aligned}$$

Solving this inequality for the error in the NO_2 integral in turn yields the bound cited in the text

$$s_{\int [\text{NO}_2] dy} \leq 0.04 \text{ ppm-km}.$$

APPENDIX C: CALCULATION OF PHOTOSTATIONARY NO₂

The NO₂ concentration can be calculated from more easily modeled quantities through the following algebraic identity (White, 1977)

$$\begin{aligned} 2[\text{NO}_2] &= [\text{O}_3][\text{NO}]/[\text{NO}_2] + ([\text{NO}_2] + [\text{O}_3]) \\ &\quad + [\text{NO}_x] - ([\text{O}_3][\text{NO}]/[\text{NO}_2]) \\ &\quad + ([\text{NO}_2] + [\text{O}_3]) + [\text{NO}_x])^2 \\ &\quad - 4([\text{NO}_2] + [\text{O}_3])([\text{NO}_x])^{1/2} \end{aligned}$$

At photostationary state (Leighton, 1961) the term $[\text{O}_3][\text{NO}]/[\text{NO}_2]$ is a constant, equal to the ratio k_2/k_1 of backward and forward rate constants. Following Johnson *et al.* (1980), the calculations for Figs 10 and 13 used the values $k_1 = 0.44 \text{ ppm}^{-1} \text{ s}^{-1}$ (Leighton, 1961) and $k_2 = 0.01 \exp(-0.38/\cos Z_s) \text{ s}^{-1}$ (Davis *et al.*, 1974), where Z_s is the solar zenith angle. The oxidant concentration $[\text{NO}_2] + [\text{O}_3] \approx [\text{NO}_2] + [\text{O}_3] + [\text{O}]$ is an invariant of reactions R1-R3, whether or not the system is at photostationary state. The calculations for Figs 10 and 13 assumed no NO_x in the

background air, 5% NO₂ in the stack NO_x, and a constant ozone background $[\text{O}_3]_{\text{bg}}$, in which case $[\text{NO}_2] + [\text{O}_3] = 0.05[\text{NO}_x] + [\text{O}_3]_{\text{bg}}$. Substitution of this equality in the initial identity yields an expression for $[\text{NO}_2]$ as an explicit function of $[\text{NO}_x]$

$$\begin{aligned} [\text{NO}_2] &= 0.5(k_2/k_1 + [\text{O}_3]_{\text{bg}} + 1.05[\text{NO}_x]) \\ &\quad - ((k_2/k_1 + [\text{O}_3]_{\text{bg}} + 1.05[\text{NO}_x])^2 \\ &\quad - 4([\text{O}_3]_{\text{bg}} + 0.05[\text{NO}_x])([\text{NO}_x])^{1/2}) \\ &= F([\text{NO}_x]; k_2/k_1, [\text{O}_3]_{\text{bg}}) \end{aligned}$$

The NO₂ integral across a Gaussian NO_x plume can thus be written

$$\int [\text{NO}_2] dy = \int F(c \exp(y^2/2\sigma_y^2); k_2/k_1, [\text{O}_3]_{\text{bg}}) dy,$$

where c is the centerline concentration of NO_x and σ_y is the dispersion parameter for the NO_x plume. In the calculations for Figs 10 and 13, c was taken to be $[\text{NO}_x]_{\text{max}}$, the maximum NO_x concentration recorded on the traverse. The dispersion parameter was defined by the relationship $\int [\text{NO}_x] dy = \sqrt{2\pi} \sigma_y [\text{NO}_x]_{\text{max}}$, which is an identity for Gaussian plumes.